

PRELIMINARY REMARKS

Claims 1 to 6 and 8 to 14 as set forth in Appendix II of this paper are now pending in this case. Claim 7 has been canceled and new Claims 8 to 14 have been added, as indicated in Appendix I of this paper.

New Claims 8 to 12 essentially correspond to Claims 1 to 5 with the difference that the presence of an alkali metal as a dopant is mandatory in the catalysts (a) to (c). New Claim 13 relates to the process wherein cesium is used as the alkali metal in accordance with applicants' disclosure on page 4, indicated lines 15 to 34, of the application and the examples disclosed in the application. New Claim 14 corresponds to Claim 7 with the difference that some editorial changes were made in the claim language. No new matter has been added.

The Examiner has rejected Claims 1 to 7 under 35 U.S.C. §103(a) as being unpatentable in light of the teaching of *Blechschnitt et al.* (US 4,077,984) or *Ueda et al.* (US 5,229,527). Favorable reconsideration of the Examiner's position and withdrawal of the rejection under Section 103 is respectfully solicited. It is respectfully urged that neither the disclosure of *Blechschnitt et al.* nor the teaching of *Ueda et al.*, each taken alone, nor a combination of *Blechschnitt et al.*'s and *Ueda et al.*'s disclosure renders the invention as a whole which is defined in applicants' independent claims prima facie obvious.

The disclosure of *Blechschnitt et al.* relates to a catalytic process for manufacturing phthalic anhydride in a high yield and high purity from a feed composition which has a high hydrocarbon content (ie. col. 1, indicated lines 60 to 64, of US 4,077,984). As explained by *Blechschnitt et al.*, excessively active catalysts only tolerate a low content of hydrocarbon in the carrier gas and therefore provide the phthalic anhydride only in poor yields, whereas catalysts which tolerate a high content of hydrocarbon and which provide the phthalic anhydride in good yields are problematic because of the poor quality of the product, ie. the heavy contamination of the product with undesirable by-products (ie. col. 1, indicated lines 54 to 59, of US 4,077,984). In accordance with the teaching of *Blechschnitt et al.* the problem can be addressed by utilizing two different catalysts, namely

- (1) a first catalyst, which accounts for from 25 to 50%-vol. (based on the total catalyst volume), and contains from 0.01 to 0.3%-wt. (based on the TiO_2 content of the catalyst) of rubidium, but no phosphorous, in the active composition, and
- (2) a second catalyst, which accounts for the remainder of the catalyst volume, and contains from 0.02 to 0.8%-wt. (based on the TiO_2 content of the catalyst) of phosphorous, but no rubidium, in the active composition,

arranged so that the hydrocarbon/carrier gas feed first gets into contact with the first catalyst (ie. col. 1, indicated line 65, to col. 2, indicated line 13, of US 4,077,984). **Blechschnitt et al.** teach that the particularities of the catalysts and their arrangement allow for the oxidation of feed compositions comprising up to 100 g/m³, preferably 44 to 80 g/m³, of hydrocarbon (ie. col. 3, indicated lines 34 to 42, of US 4,077,984). As shown in the examples and the comparative experiments provided by **Blechschnitt et al.**, the purity of the recovered phthalic anhydride is distinctly impaired if the rubidium free catalyst layer (2) is omitted. The data also show that the contamination of the product increases when the loading is increased (ie. the data compiled in the tables col. 4 of US 4,077,984). Further, as shown by applicants' investigations, two-layered catalysts do not allow for an increase in the loading beyond 100 g/Nm³.

In accordance with applicants' invention, the process is conducted in the presence of at least three catalysts, and the three catalysts are arranged in superimposed zones. Furthermore, in accordance with applicants' invention, each of the catalyst layers may contain phosphorous and each of the layers may contain alkali metal. Applicants have found that the utilization of the particular layered catalyst arrangement defined in the claims provides for a number of distinct advantages in conducting the process which advantages are not achieved by two-layered catalyst arrangements:

Firstly, due to the particularities of the layered catalyst structure, applicants' process can be conducted at loadings which are higher than 100 g/Nm³. The respective advantage is, for example, corroborated by applicants' data concerning Examples 14 and 15, and the results of the comparative investigations into two-layered structures in Examples 16 and 17¹⁾. The pertinent data are compiled in the fol-

1) The data which are reported in connection with the Examples in the specification are summarized in the table set forth in Appendix III to allow an easier overview.

lowing table (for a complete compilation of the data set forth in applicants' examples see the table in Annex III):

Example No.	Catalyst No. (Layer)	Cs [%]	Active Composition [%]	Loading [g/Nm ³]	Running-up Time [d]	Yield PSA [weight-%]	Yield PHD [weight-%]
14	I (a)	0.40	7.1	105	13	110	0.14-0.19
	IV (b)	0.40	10.5				
	VII (c)	0.10	10.5				
15	II (a)	0.35	7.5	105	15	111.5	0.14-0.24
	V (b)	0.35	10.1				
	VII (c)	0.10	10.5				
16	IV (b)	0.40	10.5	105	/	/	/
	VII (c)	0.10	10.5				
17	I (a)	0.40	7.1	105	/	/	/
	VII (c)	0.10	10.5				

The data concerning Examples 14 and 15 show that applicants' process wherein the particular, at least three-layered catalyst combination is used proceeded well even at loadings of 105 g/Nm³. In the presence of the corresponding two layered catalyst used in Example 16, the loading could not be increased beyond values of 90 g/Nm³ (ie. applicants' remarks in Table 2, col. 3, page 14 of the application). In the presence of the corresponding two layered catalyst used in Example 17, the loading could be increased to the desired value of 105 g/Nm³. However, the product was contaminated by high amounts of phthalide (PHD) and significant amounts of xylene once the loading reached 90 g/Nm³ (ie. applicants' remarks in Table 2, col. 3, page 14 of the application).

Different loadings of the process, for example the differences between loadings of 60 g/Nm³, 80 g/Nm³, and 100 g/Nm³, have a considerable impact on the economy of the process. A 15000 tube reactor providing PSA in a yield of 114% provides,

- at a loading of 60 g/Nm³ 0.7 t PSA per hour,
- at a loading of 80 g/Nm³ 0.93 t PSA per hour, and
- at a loading of 100 g/Nm³ 1.16 t PSA per hour.

The calculation shows that an increase in the tolerable or achievable loading by 20% correlates to an increase in the recoverable yield per hour by 20%.

The teaching of *Blechschnitt et al.* neither suggests nor implies that the amount of layers in which the catalysts are arranged have an

impact on the tolerable or achievable loading²⁾. Accordingly, the teaching of *Bleichschmitt et al.* contains nothing which renders this aspect of applicants' invention *prim facie* obvious.

Secondly, the particularities of the layered catalyst structure provide that applicants' process requires short periods for running-up and yields in a product which exhibits reduced contamination. This particular advantage is, for example, corroborated by applicants' data concerning Examples 8 and 9, and the results of the comparative investigations into two-layered structures in Examples 10 and 11. The pertinent data are compiled in the following table:

Example No.	Catalyst No. (Layer)	Cs [%]	Active Composition [%]	Loading [g/Nm ³]	Running-up Time [d]	Yield PSA [weight-%]	Yield PHD [weight-%]
8	I (a)	0.40	7.1	80	7	111.5	0.1-0.19
	IV (b)	0.40	10.5				
	VII (c)	0.10	10.5				
9	II (a)	0.35	7.5	80	10	113	0.15-0.25
	V (b)	0.35	10.1				
	VII (c)	0.10	10.5				
10	IV (b)	0.40	10.5	80	32	112.5	0.05-0.22
	VII (c)	0.10	10.5				
11	I (a)	0.40	7.1	80	10	113	0.37-0.58
	VII (c)	0.10	10.5				

The data concerning loadings of 80 g/Nm³ show that in accordance with applicants' process the running-up times range from 7 to 10 days and the contamination of the PSA product by phthalid (PHD) ranges from 0.1 to 0.25 weight-%. When the catalyst is only two-layered, however, either the running-up time increases to 32 days (*Example 10*), or the contamination of the PSA product by phthalid (PHD) increases to from 0.37 to 0.58 weight-% (*Example 11*).

The time interval which is required for running-up the process have an immediate impact on the economy of the procedure because of the increase expenditure due to the prolonged time, and due to losses in yield during running-up. An increased contamination of the product by the phthalide is equally undesirable because the phthalide cannot easily be separated from the phthalic anhydride and the presence of increased amounts of the phthalide in the phthalic anhydride reduces the quality of the product.

2) For example, the section in col 2, indicated lines 48 to 59, of *US 4,077,984*.

The teaching of *Blechschnitt et al.* neither suggests nor implies that the amount of layers in which the catalysts are arranged have an impact on the times which are necessary to run-up the process, or on the amount of the contamination. In accordance with *Blechschnitt et al.*'s process the contamination is influenced by the nature of the layers of catalysts. The statements in col 2, indicated lines 48 to 59, of *US 4,077,984*, indicate that *Blechschnitt et al.* consider the two-layered arrangement of their catalyst compositions to exhibit the same or similar properties as an arrangement wherein each of the two catalyst layers consists of two or more sub-layers. *Blechschnitt et al.*'s disclosure also contains no indication that the number of distinct layers could affect the time which is required to run-up the process. Accordingly, the teaching of *Blechschnitt et al.* contains nothing which renders these aspects of applicants' invention *prima facie* obvious.

It is well established that the inventive subject matter as a whole, which is referred to in the statute, embraces not only the subject matter particularly recited in the claims, but also the properties which are inherent in the particular combination of features defined in the claims, as well as the problem which is solved³). Since *Blechschnitt et al.*'s disclosure fails to teach or suggest how to improve on the run-up times and the loading values of the process, and -at the same time- to reduce the problems incurred in phthalid contamination of the product, the teaching of *Blechschnitt et al.* fails to render applicants' invention as a whole *prima facie* obvious. Favorable reconsideration of the Examiner's position and withdrawal of the rejection under Section 103(a) based on the disclosure of *Blechschnitt et al.* is, therefore, respectfully solicited.

The teaching of *Ueda et al.* relates to a process which is particularly adapted for converting a mixture of o-xylene and naphthalene into phthalic anhydride. In accordance with *Ueda et al.*'s disclosure, the process is conducted in the presence of two catalyst layers, wherein one of the layers is specifically adapted to catalyze the reaction of o-xylene and a second layer is specifically adapted to

3) ie. *In re Antonie*, 559 F.2d 618, 195 USPQ 6 (CCPA 1977); *In re Wright*, 848 F.2d 1216, 6 USPQ2d 1959 (Fed. Cir. 1988), overruled on other grounds in *In re Dillon*, 919 F.2d 688, 16 USPQ2d 1897 (Fed. Cir. 1990) (*en banc*), cert. denied 500 U.S. 904 (1991)

catalyze the reaction of naphthalene⁴). Accordingly, while the first layer aiming at the conversion of o-xylene is inadequate to provide for satisfactory results in the conversion of naphthalene, the inadequacies are obviated by the second layer. By the same token, the first layer obviates the inadequacy of the second layer for the conversion of o-xylene. *Ueda et al.*'s teaching therefore provides for a particular arrangement of the two catalysts and a correlation of the height of each of the catalyst layers to allow that the mixing ratio of o-xylene and naphthalene can be varied broadly. Since each of the catalysts utilized in the process disclosed by *Ueda et al.* aims at one of the two distinct components of the mixture, the inclusion of a third or further layers of the catalyst bed is neither suggested nor implied.

Furthermore, the disclosure of *Ueda et al.* does not address problems which are due to the time which is required for running-up the process, or suggests or implies that the amount of catalyst layers has an impact on the loading capacity or the degree to which the phthalic anhydride is contaminated by phthalide. When applicants' invention is considered as a whole, as appropriate under Section 103(a), the teaching of *Ueda et al.* can therefore not be considered as rendering it *prima facie* obvious⁵). Favorable reconsideration of the Examiner's position and withdrawal of the respective rejection is, therefore, respectfully solicited.

Neither the disclosure of *Blechschnitt et al.* nor the teaching of *Ueda et al.* correlate the number of catalyst layers with factors such as the loading capacity or the running-up time. A person of ordinary skill in the art would therefore not have been motivated to improve on such factors by varying that number. Even when the teachings of the respective references are regarded together rather than separately, the motivation to make the modification which is necessary to arrive at applicants' invention is lacking. Even under those circumstances, applicants' invention is therefore not rendered obvious under Section 103(a). Favorable action is respectfully solicited.

4) Note, for example, col. 2, indicated lines 39 to 61, in conjunction with col. 4, indicated lines 14 to 28, of *US 5,229,527*.

5) See also the criteria necessary to establish obviousness under Section 103(a) which are explained in MPEP §2142.

REQUEST FOR EXTENSION OF TIME:

It is respectfully requested that a three month extension of time be granted in this case. A check for the \$920.00 fee is attached.

Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees to Deposit Account No. 11.0345. Please credit any excess fees to such deposit account.

Respectfully submitted,

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Encl.: THE CHANGES IN THE CLAIMS (Appendix I)
THE AMENDED CLAIMS (Appendix II)
COMPILATION OF THE PERTINENT DATA (Appendix III)

HBK/BAS

A P P E N D I X I:

THE CHANGES IN THE CLAIMS (version with markings, showing the changes made):

Claim 7 has been canceled. New Claims 8 to 14 have been added as follows:

- B1
8. (new) A process for preparing phthalic anhydride by catalytic gas-phase oxidation of xylene and/or naphthalene by a gas comprising molecular oxygen in a fixed bed at elevated temperature, which process comprises conducting the oxidation in the presence of at least three coated catalysts arranged in superposed zones, which catalysts have a layer of a catalytically active composition of metal oxides applied to a core of nonporous support material, the active composition comprising from 3 to 8% by weight of V_2O_5 , from 0 to 3.5% by weight of Sb_2O_3 , from 0 to 0.3% by weight of P, alkali metal selected from the group consisting of potassium, rubidium and cesium as a dopant, and as balance TiO_2 in anatase form having a BET surface area of from 18 to 22 m^2/g , wherein the catalyst activity rises from zone to zone from the gas inlet end to the gas outlet end and the activity of the catalysts of the individual zones is set such that
- a) the least active catalyst comprises from 5 to 9% by weight, based on the total catalyst, of the active composition and the active composition comprises from 0.1 to 0.5% by weight of alkali metal (calculated as metal);
 - b) the next more active catalyst comprises from 1 to 5% by weight (absolute) more of the active composition than the catalyst (a), and the active composition comprises from 0 to 0.25% by weight (absolute) less of alkali metal than the active composition of the catalyst (a); and
 - c) the most active catalyst comprises from 1 to 5% by weight (absolute) more of the active composition than the catalyst (a), and the active composition comprises from 0.15 to 0.4% by weight (absolute) less of alkali metal than the active composition of the catalyst (a).
9. (new) The process of claim 8, wherein cesium is used in amounts of from 0.25 to 0.5% by weight as alkali metal in the catalyst (a).

- B.1*
Amended
10. (new) The process of claim 8, wherein the catalyst (c) comprises the same amount or a greater amount of active composition and less alkali metal as dopant than the catalyst (b).
 11. (new) The process of claim 8, wherein the catalyst (b) comprises a greater amount of active composition and less alkali metal as dopant than the catalyst (a).
 12. (new) The process of claim 8, wherein the catalyst (b) comprises the same amount of active composition and more alkali metal as dopant than the catalyst (c).
 13. (new) The process of claim 8, wherein the catalysts (a) to (c) comprise cesium as alkali metal.
 14. (new) The process of claim 8, wherein
 - (a) has an active composition content of from 7 to 8% by weight comprising from 6 to 8% by weight of V_2O_5 and from 0.3 to 0.4% by weight of Cs,
 - (b) has an active composition content of from 9 to 11% by weight comprising from 0.2 to 0.4% by weight of Cs, and
 - (c) has an active composition content of from 9 to 11% by weight comprising from 0.05 to 0.2% by weight of Cs.
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